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Advanced Integration

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September 12, 2018

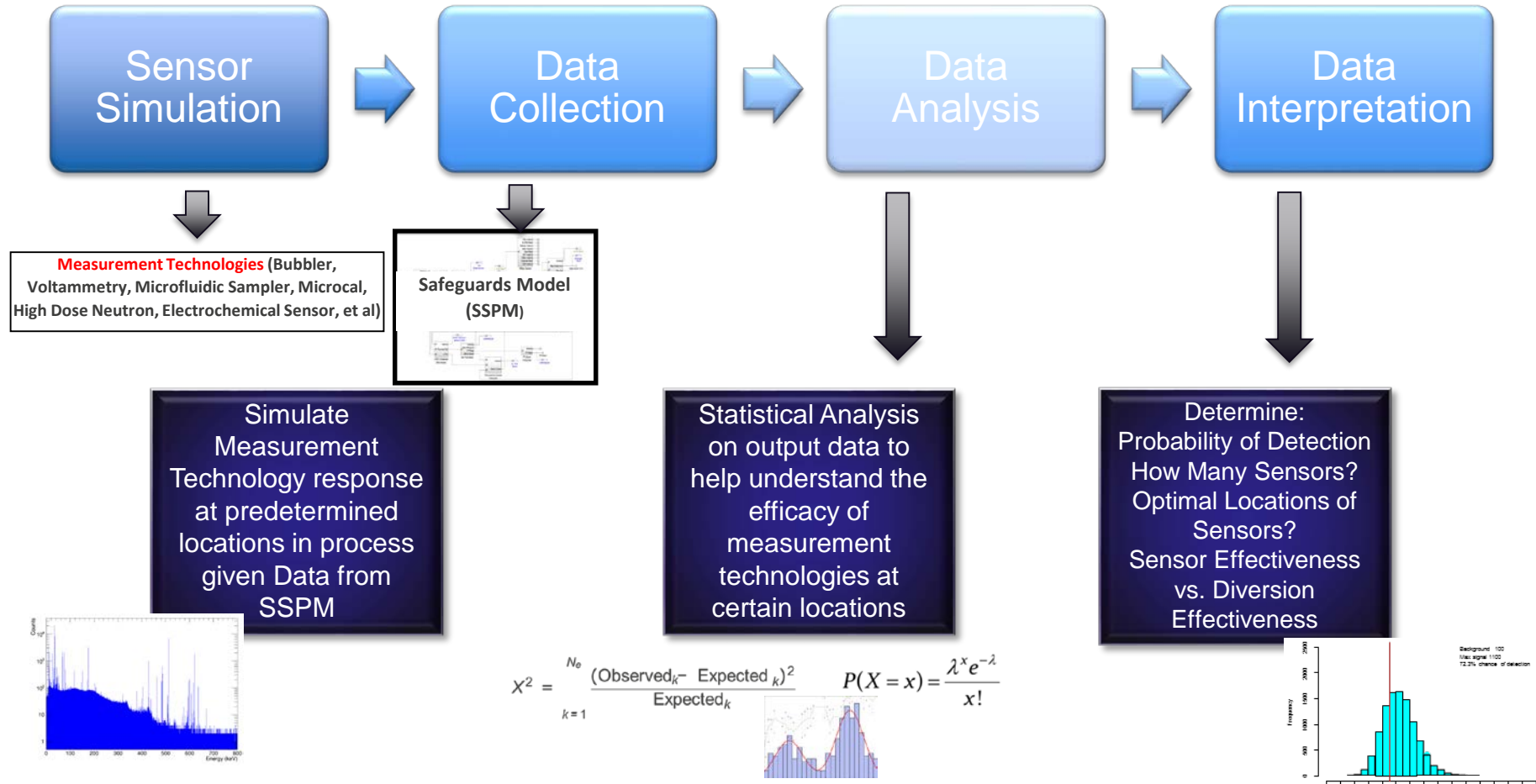


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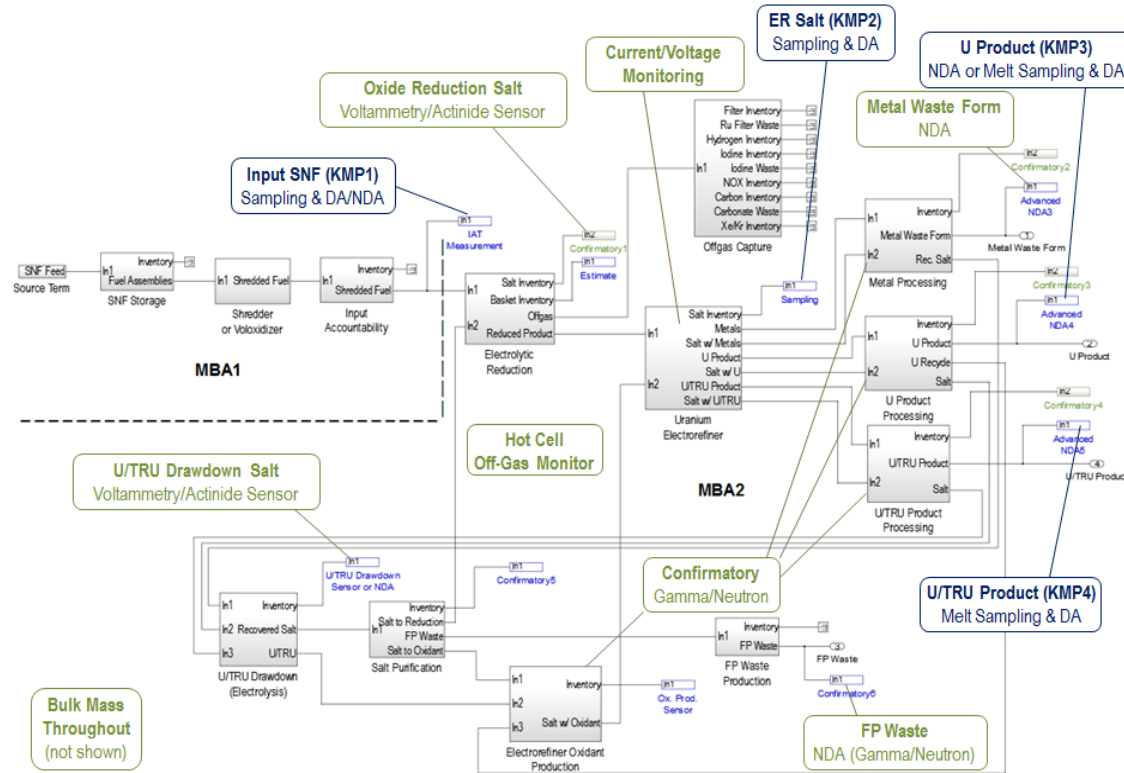
Outline

- Current Data Set being used
- Current Results for MC Modeling and Statistical Modeling of the High Dose Neutron Detector (HDND)
- Current work on new Microcal Data and Results
- Current work on Voltammetry Data

Current Framework for Advanced Integration

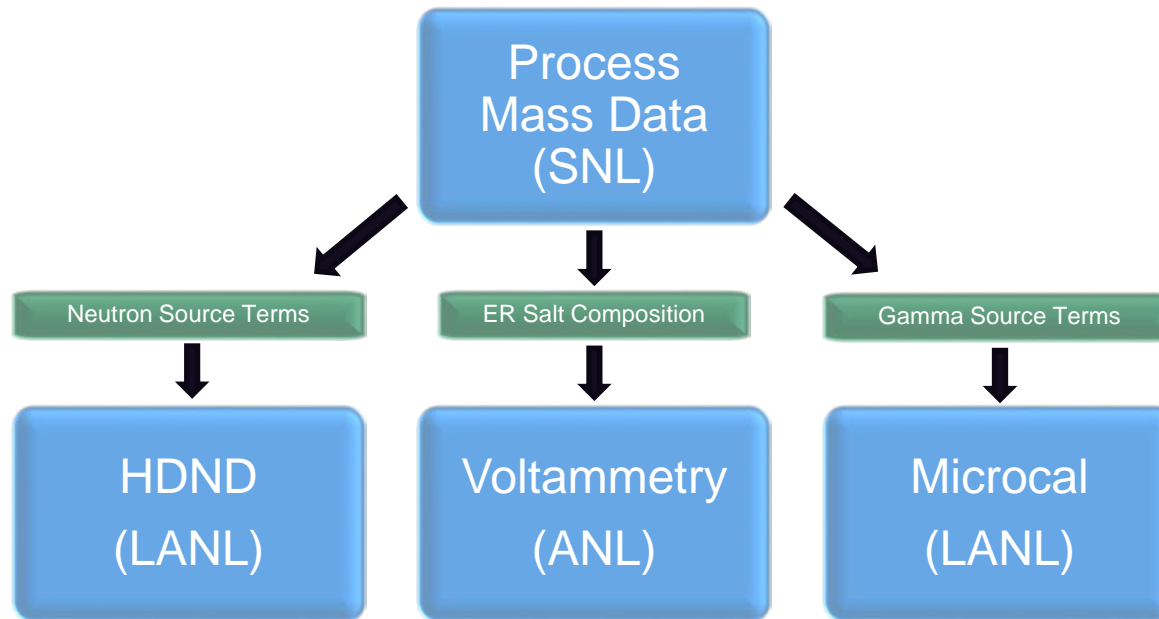


New Data Generation from SSPM – SNL, Cipiti



Data Gathering and Distribution

- Received detailed, time dependent plant isotopics from SNL
- Working with measurement technology groups to develop simulated responses based on material compositions in each process

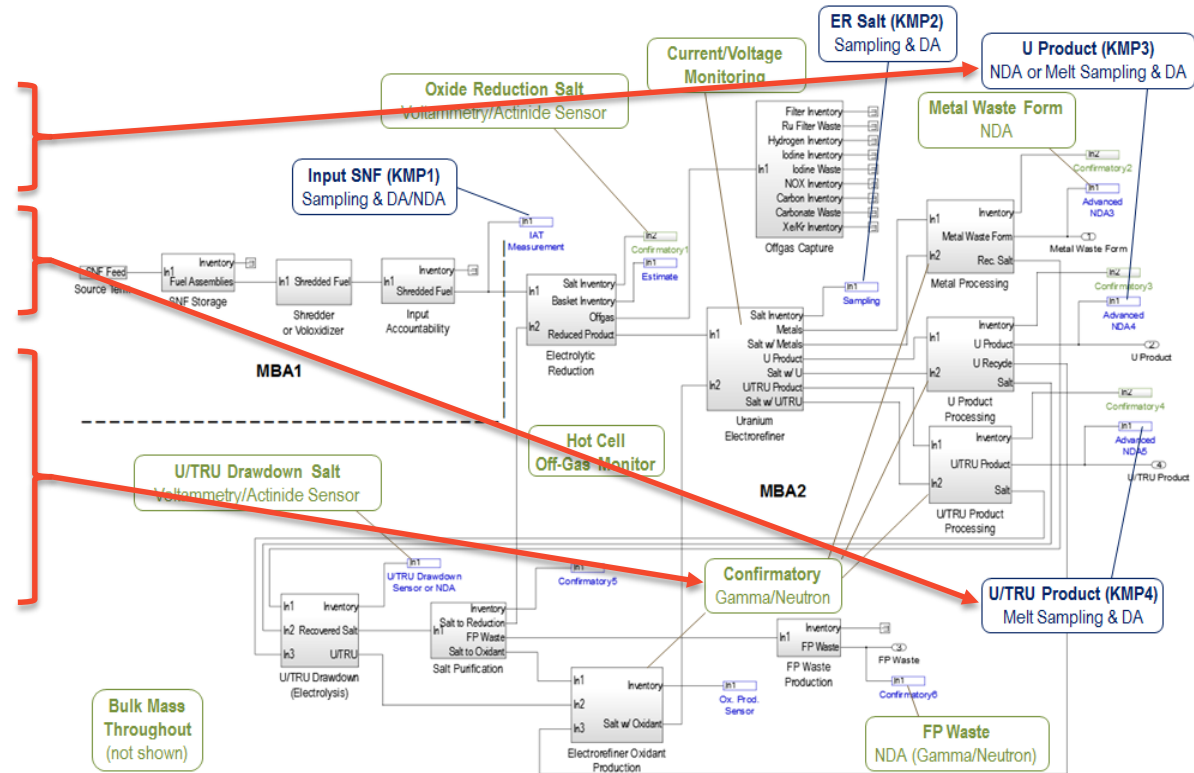


High Dose Neutron Detector (HDND)

HDND Measurement Locations

- **U Product**
 - Ensure no Pu in ingot
- **U/TRU Product**
 - Actinide accountability
- **Confirmatory Measurements**
 - Metal Processing
 - U Product Processing
 - U/TRU Product Processing
 - ER Oxidant Production

Figure from SSPM - Cipiti



U Product Neutron Signatures

- **Masses and isotopics from SSPM (Cipiti)**
- **Sources of neutrons**
 - Alpha-n reactions (negligible in U and U/TRU ingots)
 - Spontaneous fission (primary source of neutron emissions)
 - Multiplication in material
 - Induced fission
 - (n,xn)
- **Source terms**
 - U product in normal operation
 - U product with 1% Pu isotopics added
 - U product with 1% U/TRU product isotopics added

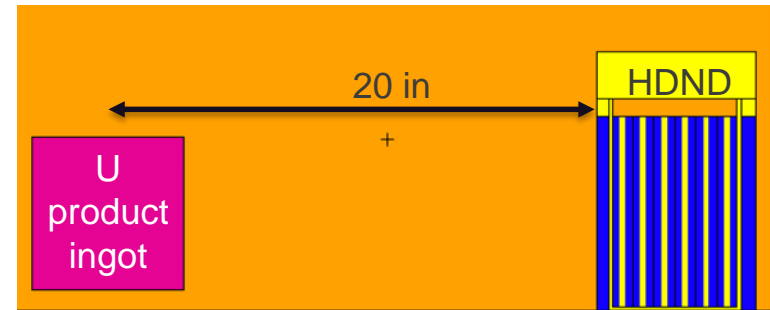
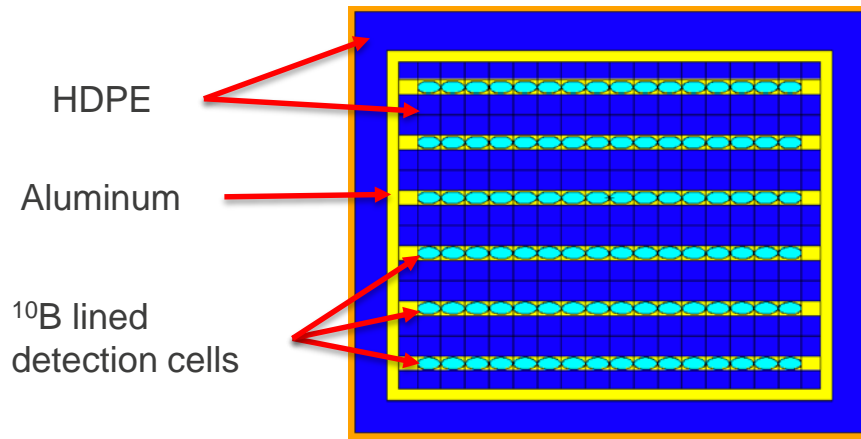
U Product
Isotopics

	U Product (n/s-cm ³)	1% Pu (n/s-cm ³)	1% U/TRU (n/s-cm ³)
U-238	0.254	0.254	0.254
Pu-238	0.000	0.169	0.169
Pu-240	0.001	0.653	0.653
Pu-242	0.000	0.342	0.342
Cm-244	0.072	0.072	83.350
Cm-246	0.003	0.003	3.369
Total	0.330	1.493	88.420

MCNP Modeling

HDND MC Modeling

- MCNP model of HDND detector
- Use neutron source term created from isotopic data
- Represent U product ingots in model
- Count neutron captures in ^{10}B that result in an energy deposition within the C10 gas sufficient to generate a pulse
- Rate of captures gives estimate of total neutron detection efficiency



HDND Statistical Modeling

- From MC modeling we expect the following count rates for U Product and U/TRU Product

	U Product	UTRU Product
	pulses/s	pulses/s
Normal	7.9	66098.1
1% Pu	36.8	64640.4
1% UTRU	2192.6	64311.5

Count rate IF 1% Pu is diverted to U Product
No Background
-This will be the focus of statistical methods used

Green = Integration Methods Completed
Yellow = Working

- Statistical methods will use 5 minute and 10 minute count rates

Probability of Detection (U Product)

	U Product	UTRU Product
	pulses/s	pulses/s
Normal	7.9	66098.1
1% Pu	36.8	64640.4

- Neutron counts are modeled as a Poisson Distribution
- Z statistic used to detect a difference in counts
- With a diversion of Pu, observed count rate will be larger, therefore Z will be large
- For a 5% False Alarm rate:
 $Z < 1.645$ (95th quantile of std Gaussian)
- Very High Probability of detection of Pu in U Product (7.9 vs 36.8)

$$Z = \frac{\text{observed} - \text{expected}}{\sqrt{\text{expected}}}$$

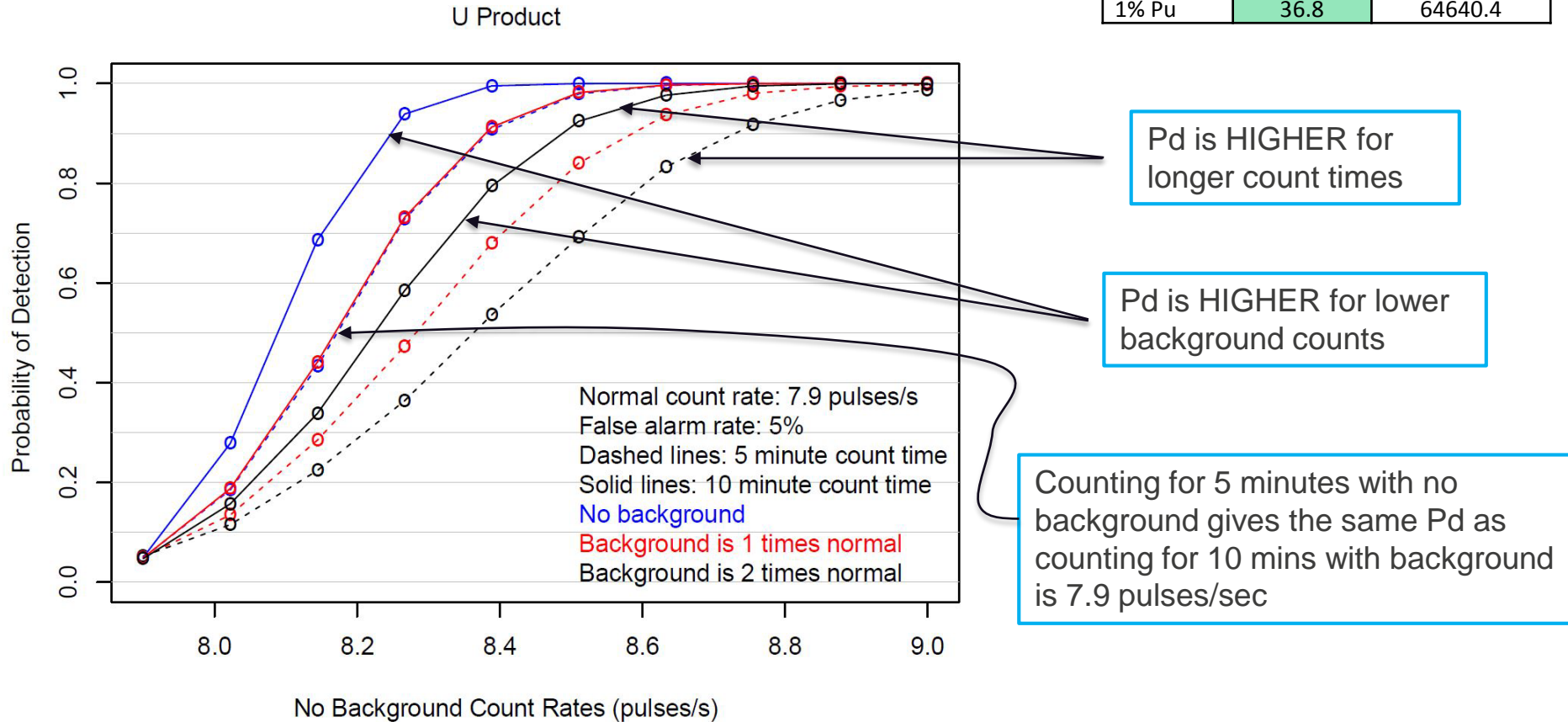
5 min count rate, 1% Pu

$$\begin{aligned}
 Z &= \frac{(5 \cdot 60 \cdot 36.8) - (5 \cdot 60 \cdot 7.9)}{\sqrt{5 \cdot 60 \cdot 7.9}} \\
 &= \sqrt{300} \cdot \frac{(36.8 - 7.9)}{\sqrt{7.9}} \approx 178.
 \end{aligned}$$

99.999th quantile of std Gaussian = 4.26

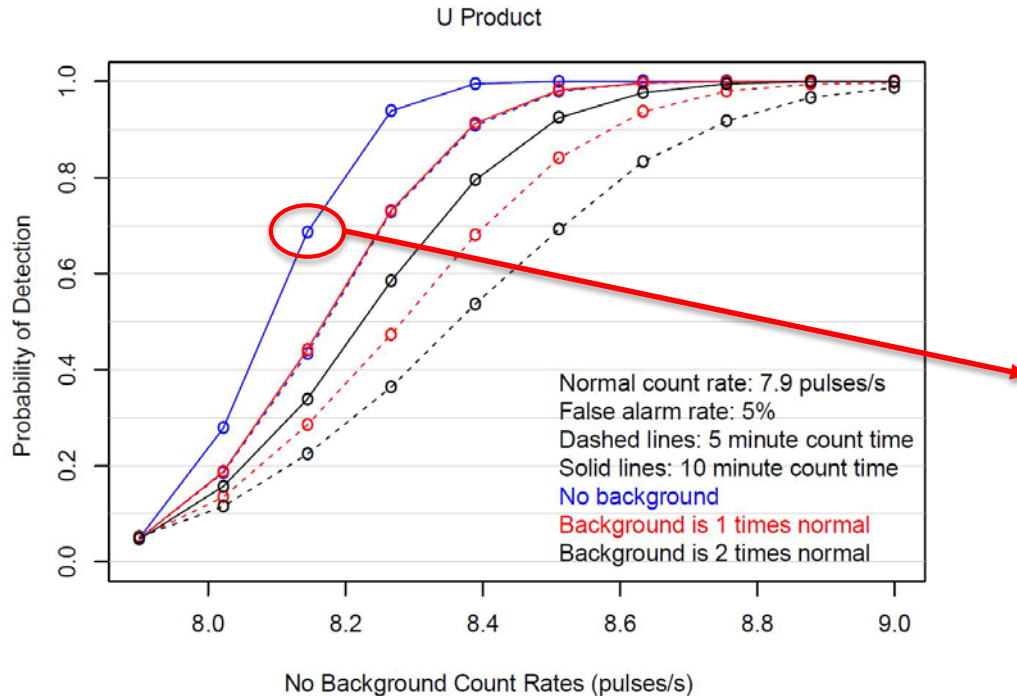
Probability of Detection (U Product)

	U Product	UTRU Product
	pulses/s	pulses/s
Normal	7.9	66098.1
1% Pu	36.8	64640.4

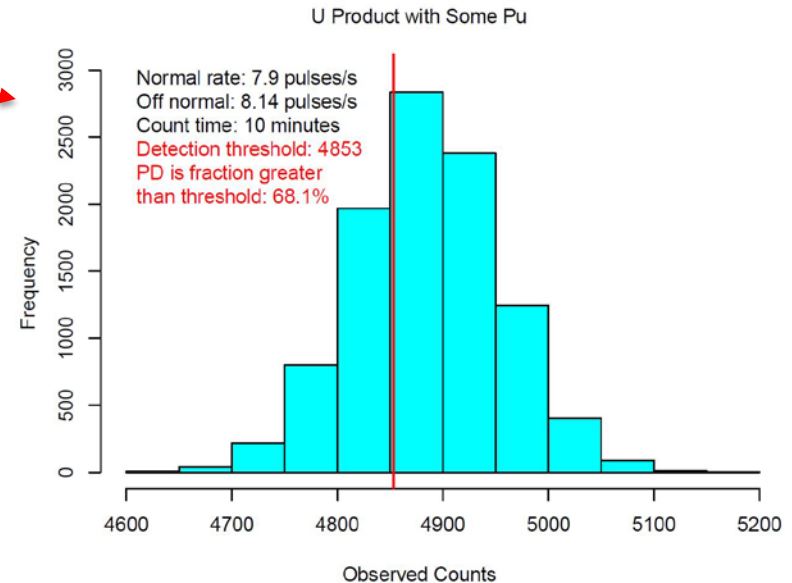


Probability of Detection (U Product)

	U Product	UTRU Product
	pulses/s	pulses/s
Normal	7.9	66098.1
1% Pu	36.8	64640.4



For every circle, Pd is estimated by counting how many times the statistic Z exceeds threshold (1.645) using 100000 Poisson Samples



Probability of Detection (UTRU Product)

	U Product	UTRU Product
	pulses/s	pulses/s
Normal	7.9	66098.1
1% Pu	36.8	64640.4

- Neutron counts go down with a diversion of Pu
- Due to counts being so high, easy to detect small decreases
- With a diversion of Pu, observed count rate will be smaller, therefore Z will be a large negative
- For a 5% False Alarm rate:
 $Z < 1.645$ (95th quantile of std Gaussian)
- Very High Probability of detection of Pu in U/TRU Product

$$Z = \frac{\text{observed} - \text{expected}}{\sqrt{\text{expected}}}$$

$$Z = \frac{(64640.4 - 66098.1)}{\sqrt{66098.1}} \sim -5.7$$

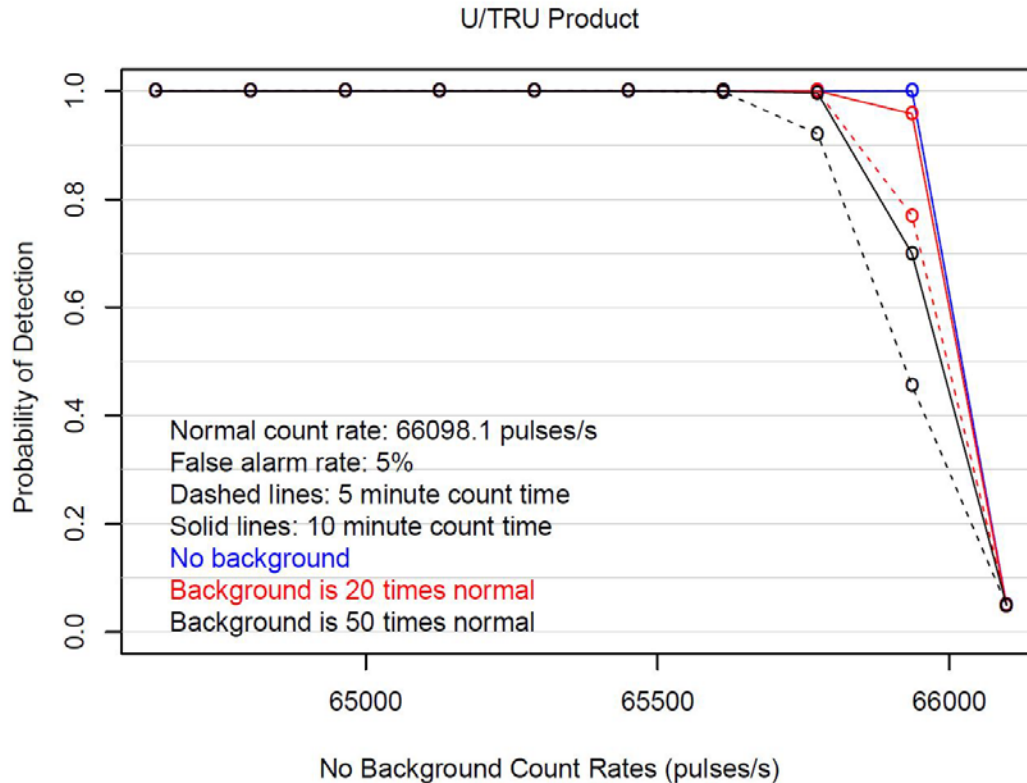
5 min count rate, 1% Pu

$$\sqrt{300}(-5.7) \sim -98$$

98 standard deviations from no diversion mean
 0.00001th quantile of std Gaussian = -4.26

Probability of Detection (UTRU Product)

	U Product	UTRU Product
	pulses/s	pulses/s
Normal	7.9	66098.1
1% Pu	36.8	64640.4



Just like U Product –
Longer count times increase Pd
Lower background increase Pd

The reason for high background rates:
Poisson data Stddev = $\sqrt{\text{mean}}$
The square root is relatively small when
counts are large

NOTE: dashed blue is
under solid red

HDND Conclusion

- The HDND has been shown to be **HIGHLY** effective at the specified locations to check U Product and U/TRU product for Pu diversions of relatively small amounts of Pu
- Efficiencies of the HDND were not considered
- HDND Simulation modeling still needs to be done for confirmatory measurements

Microcalorimeter (Microcal)

Microcalorimeter

The microcalorimeter advantage:

10X better energy resolution than semiconductor detectors

- Reduce performance gap between NDA and DA
- Reduce reliance on sampling, mass spec, and chemical analysis
- Constrain diversion scenarios
- Better resolution enables better quantitative measurements

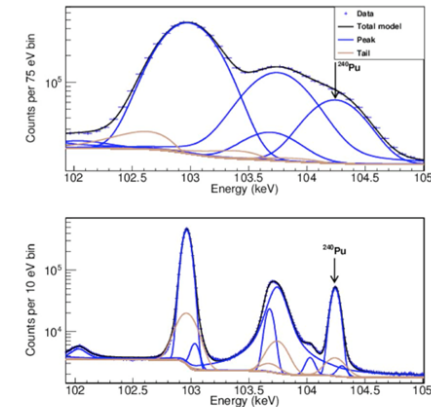
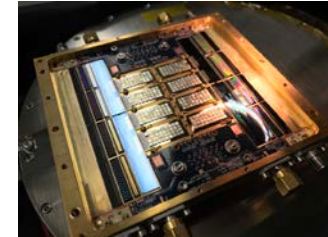
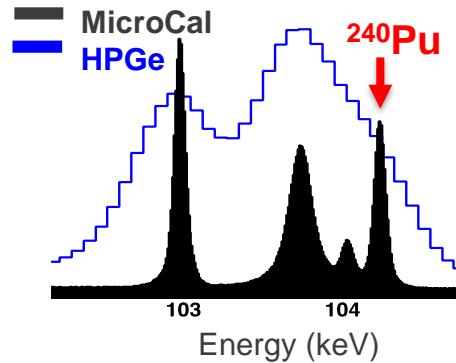
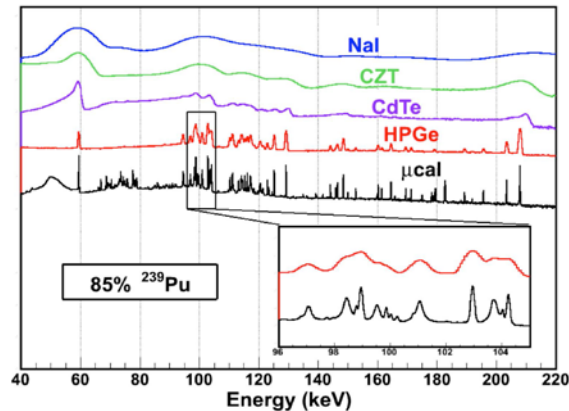


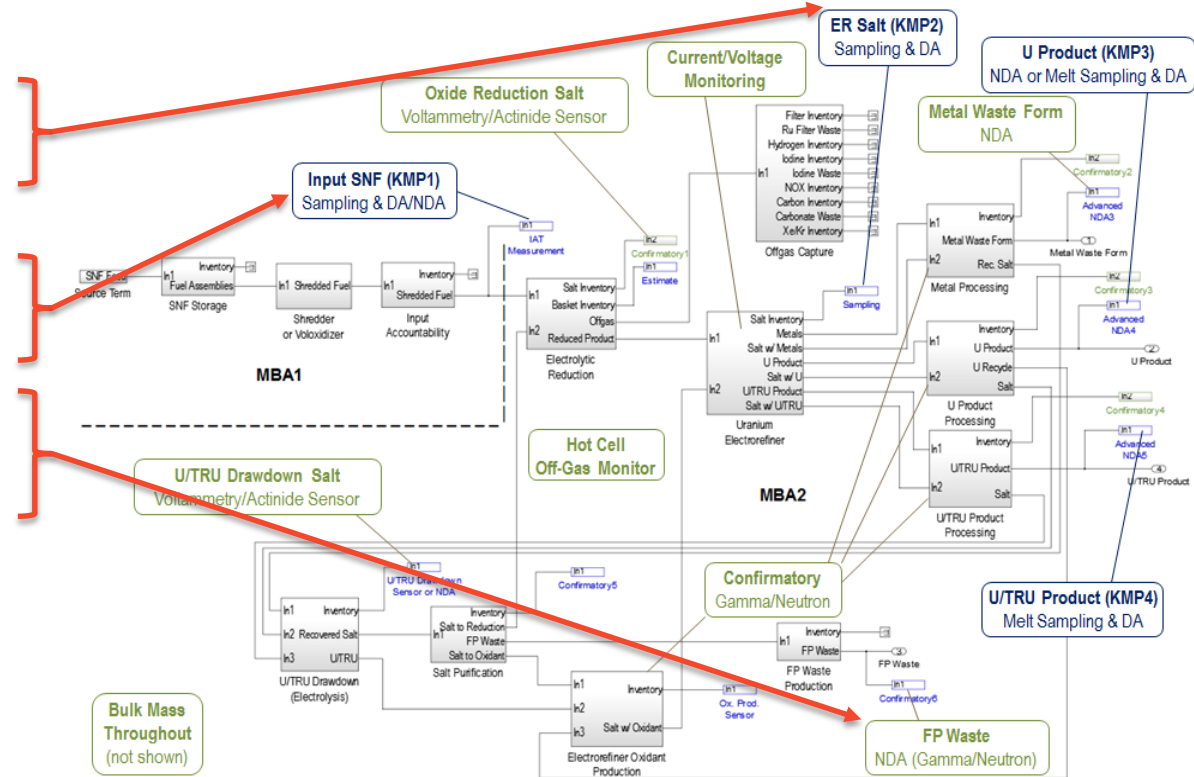
Figure 11 - Spectral region surrounding the 104.23 keV ^{240}Pu peak measured with HPGe (top) and microcalorimeter (bottom). [Hoover, 2013]

Potential applications in safeguards and material accounting:

- Spent fuel U/Pu elemental composition
- Fission products that track actinides
- Isotopic composition of U/TRU, Pu products
- Actinide content in wastes

- **ER Salt**
 - Determine the best visible peaks
- **Source**
 - Determine IF there are viable peaks
- **FP Waste**
 - For waste forms

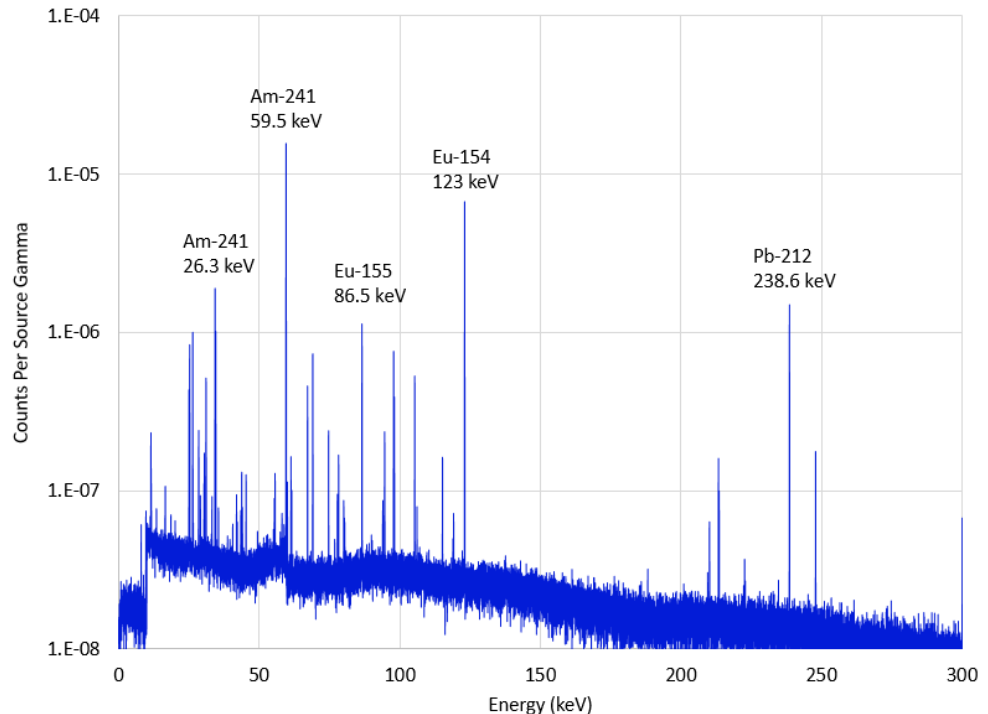
Figure from SSPM - Cipiti



Microcalorimeter

- **Gamma source terms created from mass data from SSPM measurement locations**
- **Locations evaluated**
 - Source term
 - ER salt
 - Fission Product Waste
- **Most prominent peaks belong to Am, Eu, and Pb**
- **Pu peaks difficult to identify above continuum**

ER Salt μ Cal Spectrum



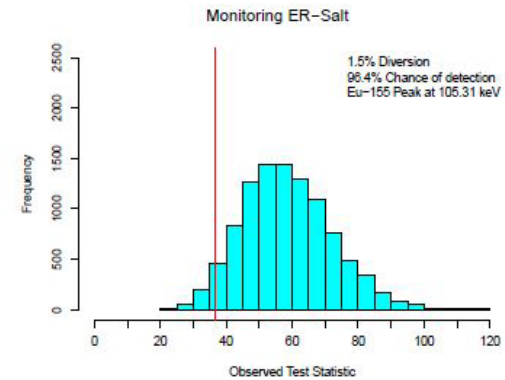
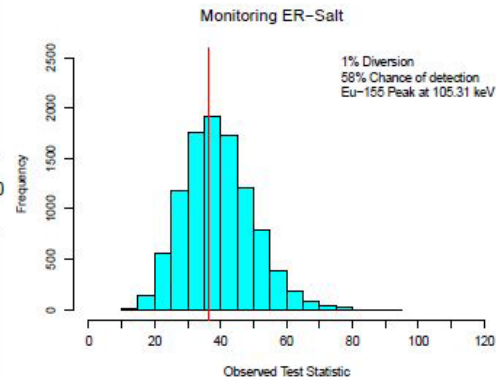
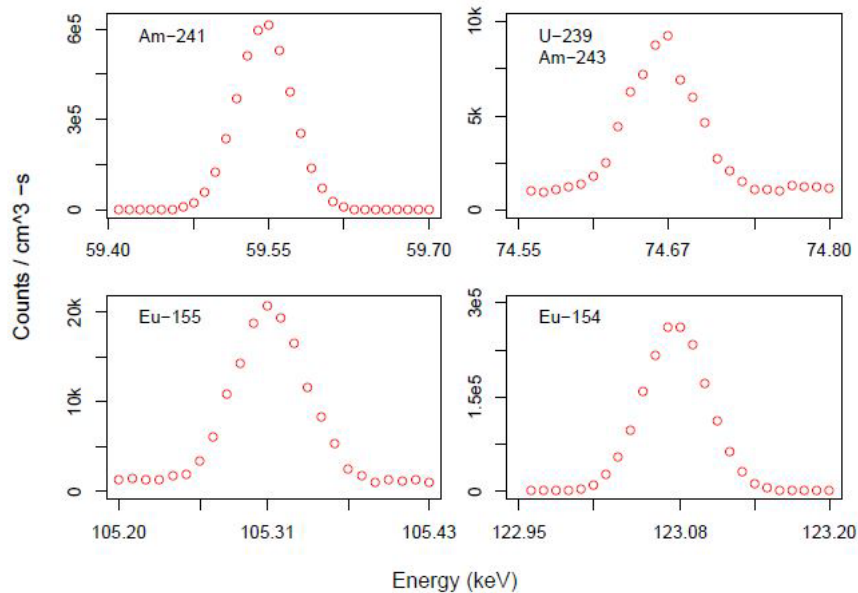
Additional Intense Microcal Peaks to consider from Simulation

ER-Salt		FP Waste		Source Term	
Energy (MeV)	Radionuclide	Energy (MeV)	Radionuclide	Energy (MeV)	Radionuclide
0.238632	Pb-212	0.238632	Pb-212	0.0595409	Am-241
0.1230706	Eu-154	0.1230706	Eu-154	0.1230706	Eu-154
0.0595409	Am-241	0.247929	Eu-154	0.238632	Pb-212
0.247929	Eu-154	0.0865479	Eu-155	0.0263446	U-237/Am-241
0.0865479	Eu-155	0.1053083	Eu-155	0.247929	Eu-154
0.1053083	Eu-155	0.115183	Pb-212	0.0865479	Eu-155
0.0263446	Am-241	0.18822	Eu-154	0.1053083	Eu-155
0.115183	Pb-212	0.045299	Eu-155	0.07466	U-239/Am-243
0.07466	U-239/Am-243	0.0600086	Eu-155	0.106123	Np-239
0.18822	Eu-154	0.176314	Sb-15	0.033196	U-237/Am-241

ER-salt isotopes of interest

Probability of Detection for 1% and 1.5% Pu Diverted, with 5% False Alarm Rate for Eu-155

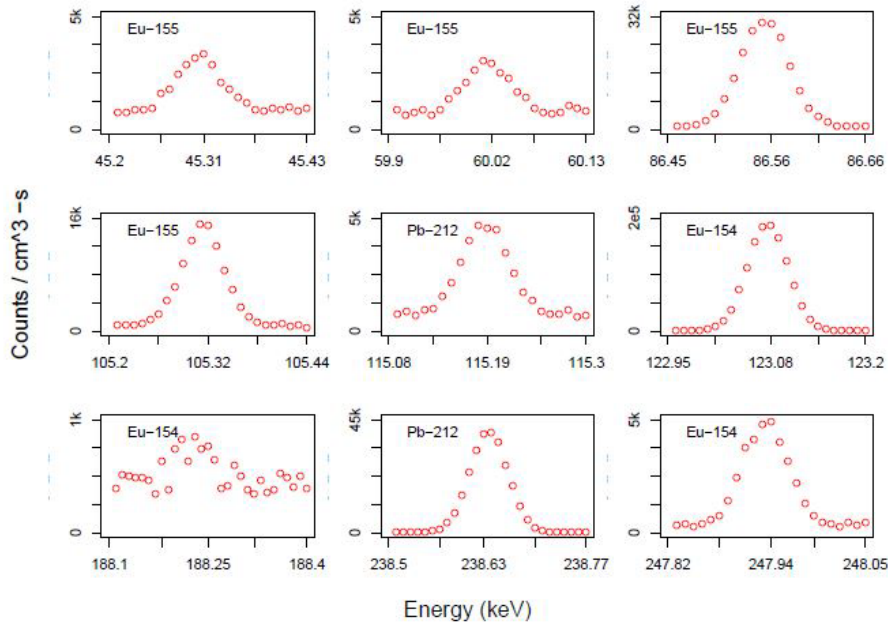
ER-Salt: Four High Intensity Peaks



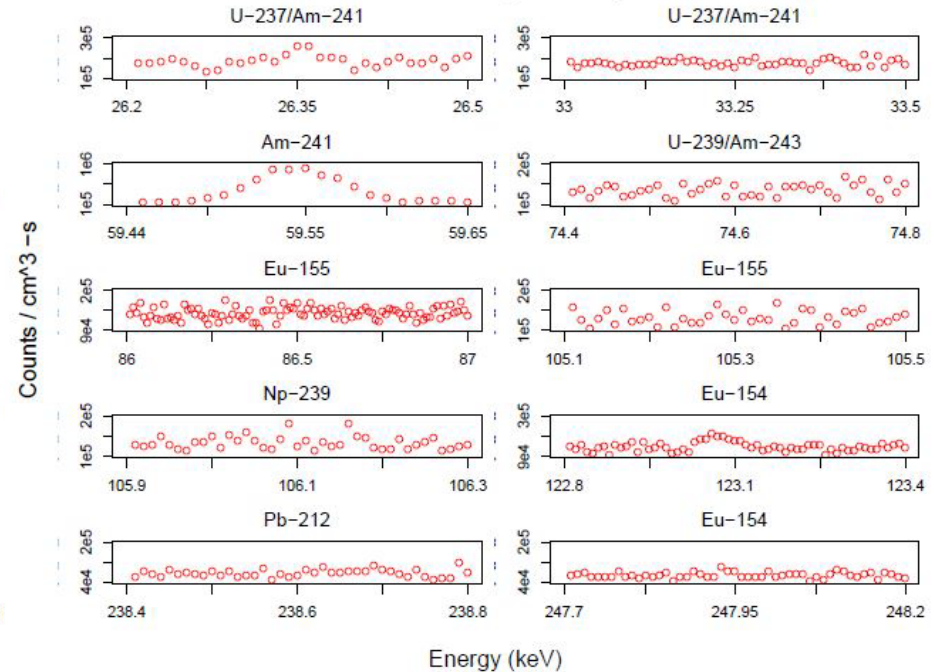
Diverted	Pd
0%	0.05
1%	0.58
1.5%	0.964

FP Waste and Source isotopes of interest

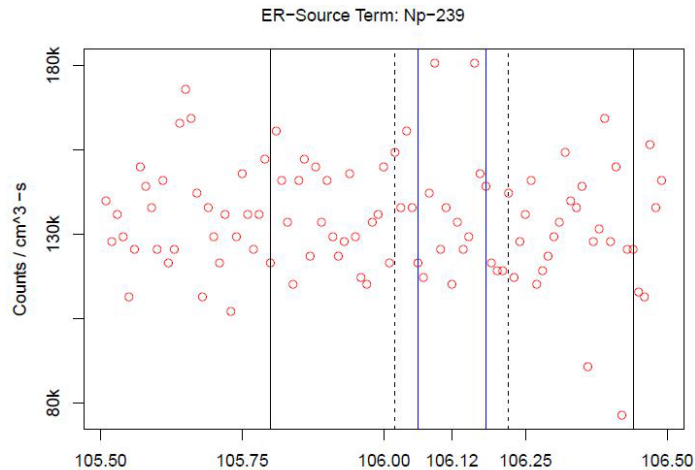
ER-FP Waste: High Intensity Peaks



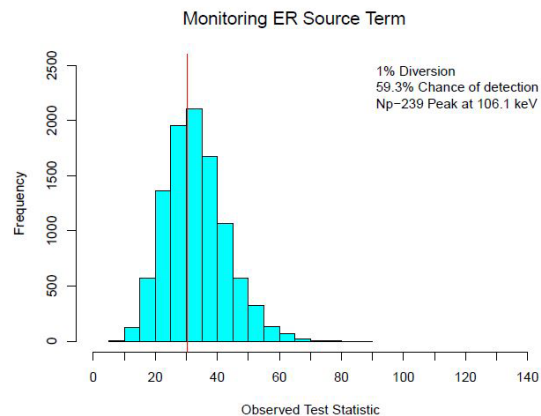
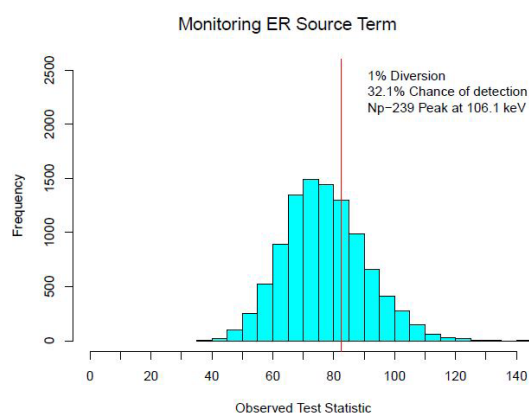
ER-Source Term: High Intensity Peaks



Source Microcal Detection



- Hard to see definitive peaks on many isotopes
- Consider Np239, peak is supposed to be at 106.12 keV.
- Noisy data points near peak lower Pd
- Its important to be able to define region of interest when determining Pd – especially when there is “noise”



Energy Bin Range	Pd (1% Diversion)
105.8 - 106.44 (solid black lines)	0.321
106.02 - 106.22 (dashed black lines)	0.593

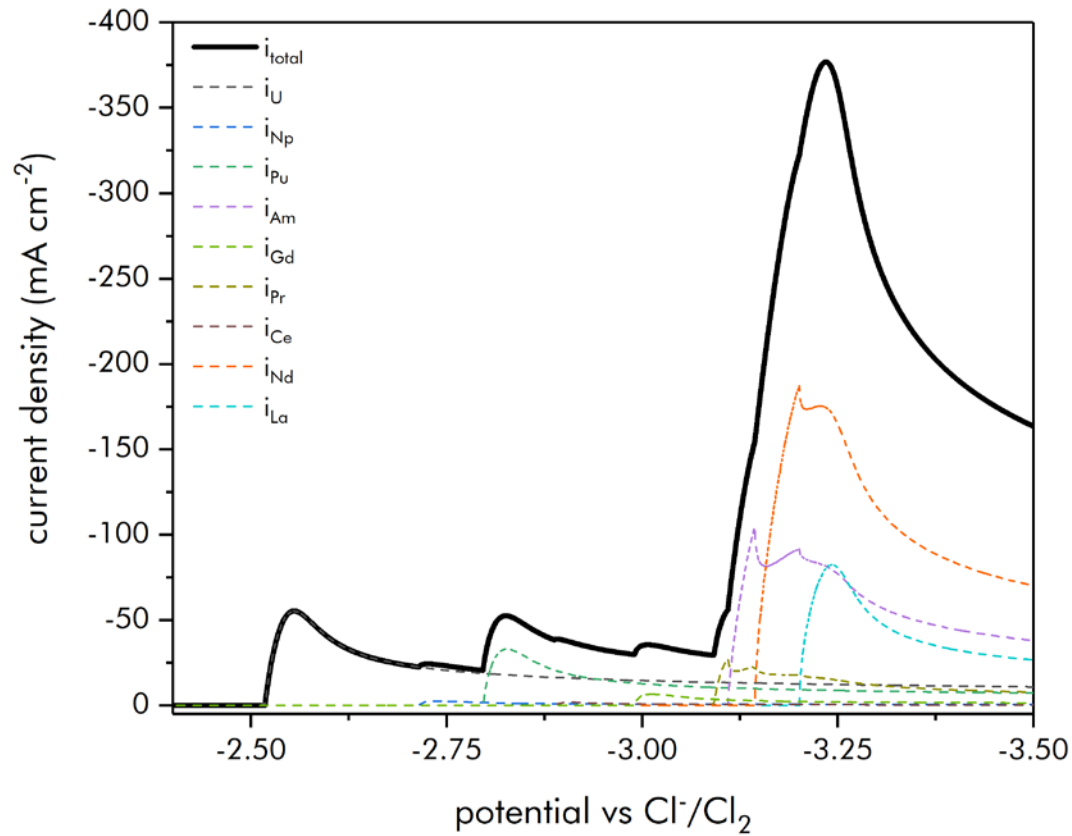
Microcal Conclusions

- Microcalorimeter is exceptional at detecting small diversions using intense peaks in the spectrum as the statistical data point
- Surrogate data for the Electro Refiner shows principle detecting elements such as Americium, Curium, Uranium, Neptunium are good indicators, **if peaks standout**
- Large peaks such as Eu-155, Am-241, Eu-154, U-239/Am-243 may be utilized for detection, once better isotopic data is known and an understanding how these isotopes changes according to diversions
- Only looked at variability in counting statistics and need to characterize other sources of uncertainty

Voltammetry

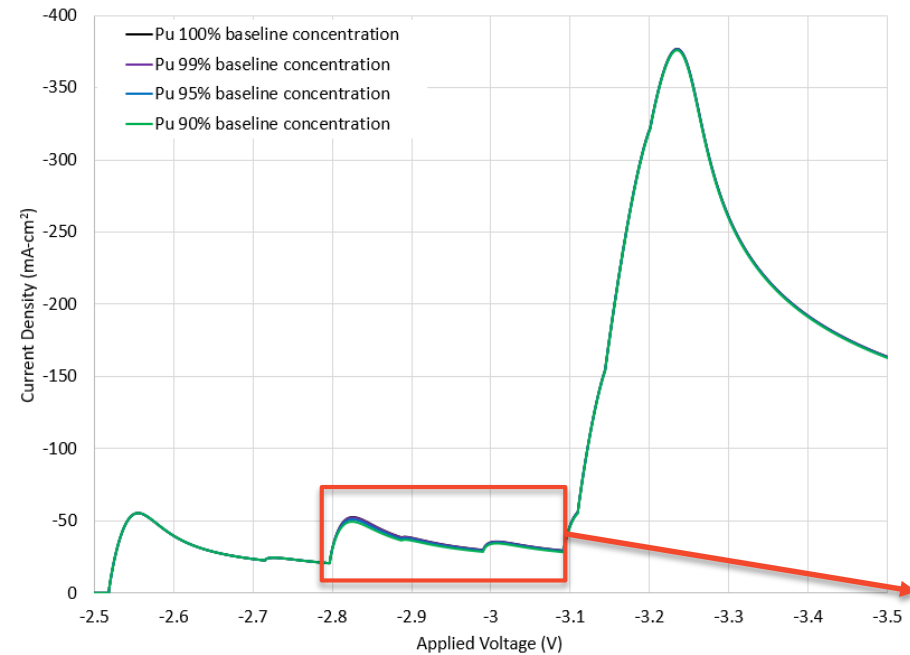
Ongoing Analysis....

Voltammetry Simulation



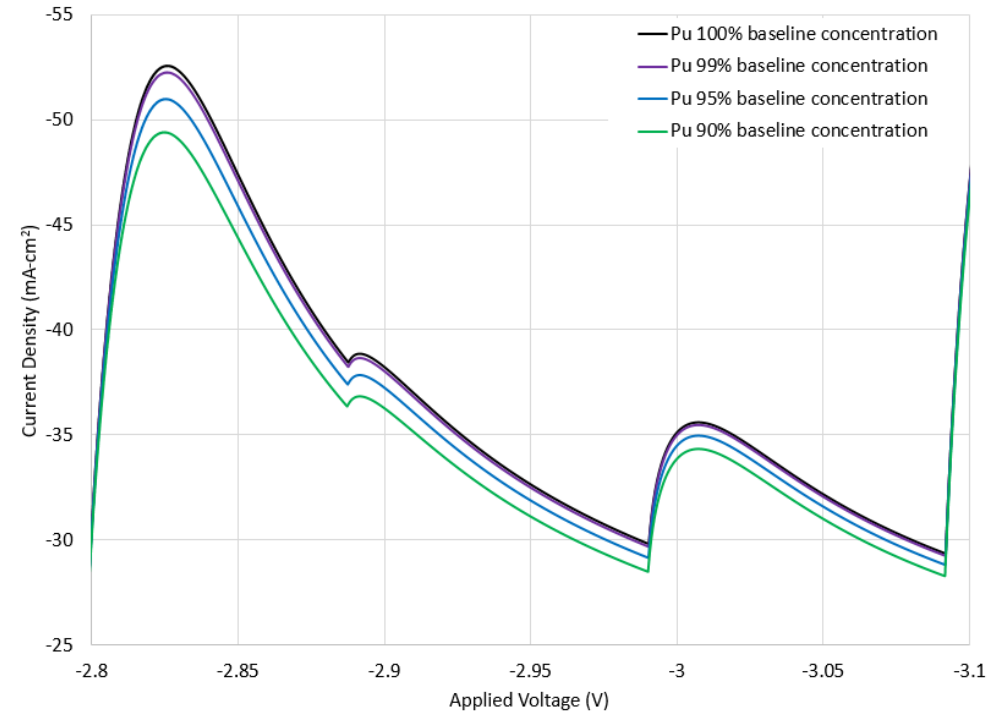
- Elemental mass data from the SSPM EChem model sent to Voltammetry development team at ANL
- N. Hoyt simulated the response of the instrument to the provided ER salt composition
- Black line represents total current density
- Dotted lines show species contributing to current density

Voltammetry Simulation, cont.



Repeated simulation, varying the Pu concentration of the ER salt to see how the waveform changes shape.

Changes in Pu concentration alters waveform slightly



Voltammetry Conclusion

- Small change in Pu concentration alter the waveform
- Need to determine if the difference from normal to off-normal concentrations is significant enough for definitive conclusions to be made regarding diversion
- Current work is ongoing
- Thanks to ANL for simulated voltammetry response

Future Work

- Analyze Confirmatory Measurement locations with simulated HDND
- Begin Voltammetry Integration Analysis with the data we've received
- Work with Bubbler SME's to determine integration approach, simulation and analysis
- Work with Microfluidic Sampler SME's to determine integration approach, simulation and analysis
- Data Flow Sheet development – currently using SSPM data, but need to double check integration of AMPYRE, DyER –what is the campaign wide process model flowsheet?



END

Test Statistic for Comparing Spectra of Counts

$$\chi^2 = \sum_{k=1}^{N_e} \frac{(\text{Observed}_k - \text{Expected}_k)^2}{\text{Expected}_k}$$

- Compare the observed counts in a particular energy bin to the expected counts—add up over all energy bins
- Expected counts from spectrum of in-control process
- χ^2 has an approximate chi-square distribution
 - Chi-squared distributions used for “goodness of fit”
- χ^2 is large when counts are different enough—set a threshold for what is large
- Threshold determines false positive and detection rate